

What is Claimed is:

1. A covalently cross-linked polymer electrolyte, the polymer electrolyte comprising amine groups in the polymer backbone and a dissolved or dispersed metal salt therein.

2. The polymer electrolyte of claim 1 wherein said electrolyte is a continuous film, having a thickness ranging from about 50 to about 1500 microns.

3. The polymer electrolyte of claim 2 wherein said electrolyte has a specific conductivity of at least about  $10^{-4}$  S/cm at a temperature ranging from about 40°C to about 60°C.

4. The polymer electrolyte of claim 1 wherein said electrolyte has a specific conductivity of at least about  $10^{-4}$  S/cm at a temperature ranging from about 40°C to about 60°C.

5. The polymer electrolyte of claim 1 wherein said polymer comprises a linear or branched, substituted or unsubstituted poly(alkylamine).

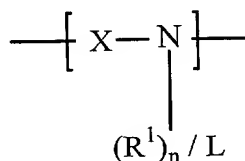
6. The polymer electrolyte of claim 5 wherein the polymer is branched.

7. The polymer electrolyte of claim 5 wherein the polymer is linear.

8. The polymer electrolyte of claim 5 wherein the polymer is selected from substituted or unsubstituted poly(ethylenimine) and substituted or unsubstituted poly(propylenimine).

9. The polymer electrolyte of claim 8 wherein the polymer is branched poly(ethylenimine).

10. The polymer electrolyte of claim 1 wherein the polymer backbone comprises a repeat unit represented by the formula:



wherein: N is nitrogen, which is optionally attached to a substituent, R<sup>1</sup>, or a covalent cross-linker, L; R<sup>1</sup> a substituent free of covalent bonds to the polymer backbone, other than the backbone containing the nitrogen atom to which it is covalently bound, and is independently selected from the group consisting of hydrogen, substituted or unsubstituted hydrocarbyl, and substituted or unsubstituted heterohydrocarbyl; L is a covalent cross-linker connecting the polymer chain to which it is attached to another polymer chain; X is the remaining portion of the repeat unit and is independently selected from the group consisting of substituted or unsubstituted hydrocarbylene, and substituted or unsubstituted heterohydrocarbylene; and, n is 1 or 2.

11. The polymer electrolyte of claim 10 wherein said polymer is a copolymer.

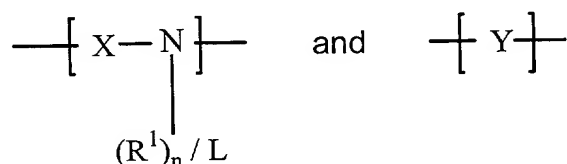
12. The polymer electrolyte of claim 11 wherein said copolymer has a backbone comprising two or more different repeat units as defined in claim 10.

13. The polymer electrolyte of claim 12 wherein X is independently selected from methylene, ethylene, propylene, butylene and pentylene.

14. The polymer electrolyte of claim 13 wherein X is ethylene and propylene, the electrolyte being a copolymer of substituted or unsubstituted, branched or linear poly(ethylenimine) and poly(propylenimine).

15. The polymer electrolyte of claim 14, wherein said copolymer is a random copolymer.

16. The polymer electrolyte of claim 10, wherein the polymer is a copolymer, the copolymer backbone comprising repeat units represented by the formulas:



5 wherein X, N, R<sup>1</sup>, n and L are as defined in claim 10, and Y is independently selected from the group consisting of substituted or unsubstituted hydrocarbylene, and substituted or unsubstituted heterohydrocarbylene.

17. The polymer electrolyte of claim 16, wherein said copolymer is a random or block copolymer.

18. The polymer electrolyte of claim 17, wherein Y is selected from substituted or unsubstituted polyethylene, polypropylene, poly(ethylene oxide), poly(propylene oxide), poly(ethylene sulfide), and poly(propylene sulfide).

19. The polymer electrolyte of claim 1, wherein the polymer is swollen with a solvent.

20. The polymer electrolyte of claim 19 wherein the solvent concentration in the electrolyte is less than about 50 weight percent, relative to the total weight of the electrolyte.

21. The polymer electrolyte of claim 19 wherein the solvent concentration in the electrolyte is less than about 25 weight percent, relative to the total weight of the electrolyte.

22. The polymer electrolyte of claim 19 wherein the solvent concentration in the electrolyte is less than about 10 weight percent, relative to the total weight of the electrolyte.

23. The swollen polymer electrolyte of claim 19 wherein the swelling solvent is selected from the group consisting of 1,2-dimethoxyethane, methyl formate, dimethylsulfoxide, sulfolane, methyl pyrrolidine, dimethyl formamide, dimethyl acetamide, glymes, nitriles, organic phosphates, organic phosphoramides, carbonates, as well as mixtures thereof.

24. The swollen polymer electrolyte of claim 23 wherein the swelling solvent is a glyme selected from the group consisting of monoglyme, diglyme, triglyme, tetraglyme, or a mixture thereof.

25. The swollen polymer electrolyte of claim 19 wherein the swelling solvent is a plasticizer.

26. The swollen polymer electrolyte of claim 25 wherein the plasticizing solvent is selected from the group consisting of esters, diesters, carbonates, phosphates, acrylates, borates, sulfolanes, sulphates and glymes.

27. The swollen polymer electrolyte of claim 26 wherein the solvent is 2-(2-ethoxyethoxy)ethyl acetate.

28. The swollen polymer electrolyte of claim 26 wherein the solvent is dimethyl adipate or dibutyl phthalate.

29. The swollen polymer electrolyte of claim 26 wherein the solvent is propylene carbonate.

30. The polymer electrolyte of claim 1, wherein the polymer is an elastomer.

31. The polymer electrolyte of claim 1 wherein a nitrogen of one or more amine groups in a first polymer backbone are covalently cross-linked to amine groups in a second polymer backbone.

32. The polymer electrolyte of claim 1 wherein the metal salt is selected from the group consisting of transition metals, alkali metals, alkaline earth metals, or a combination thereof.

33. The polymer electrolyte of claim 32 wherein the metal salt is a transition metal salt selected from the group consisting of Ni, Cu, Ru or Ag.

34. The polymer electrolyte of claim 33 wherein the metal salt is a alkali metal salt selected from the group consisting of Li, Na, K, Rb or Cs.

35. The polymer electrolyte of claim 34 wherein the metal salt is a Li metal salt selected from the group consisting of LiSCN,  $\text{LiPF}_6$ ,  $\text{LiAsF}_6$ ,  $\text{LiClO}_4$ ,  $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ ,  $\text{LiBF}_4$ ,  $\text{LiCF}_3\text{SO}_3$ ,  $\text{LiSbF}_6$ , or a combination thereof.

36. The polymer electrolyte of claim 33 wherein the metal salt is a alkaline earth metal salt selected from the group consisting of Mg, Ca or Sr.

37. The polymer electrolyte of claim 1 wherein the polymer has a ratio of secondary to tertiary nitrogen atoms ranging from about 5:1 to about 25:1.

38. The polymer electrolyte of claim 1 wherein the polymer has a ratio of secondary to tertiary nitrogen atoms ranging from about 10:1 to about 20:1.

39. The polymer electrolyte of claim 1 wherein the polymer has a ratio of heteroatoms to metal ions ranging from about 20:1 to about 4:1.

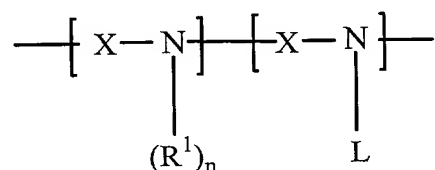
40. A covalently cross-linked polymer electrolyte, the polymer electrolyte comprising amine groups in the polymer backbone and one or more solvent moieties bound thereto.

41. The polymer electrolyte of claim 40 wherein the polymer comprises a linear or branched, substituted or unsubstituted poly(alkylamine).

42. The polymer electrolyte of claim 41 wherein the polymer is poly(ethylenimine) or poly(propylenimine).

43. The polymer electrolyte of claim 40 wherein one or more of the solvent moieties are bound to amine group nitrogen atoms.

44. The polymer electrolyte of claim 43 wherein the polymer comprises a repeat unit of the formula:



wherein: n is 1 or 2; R<sup>1</sup> is (i) free of covalent bonds to the polymer backbone other than the backbone containing the nitrogen atom to which it is covalently  
5 bound, and (ii) is a substituted or unsubstituted heterohydrocarbyl solvent moiety derived from a glyme, a furan, an amide, an alkylsulfoxide, a sulfolane,

10 a nitrile or a carbonate; L is a covalent cross-linker connecting the polymer chain to which it is attached to another polymer chain; and X is independently selected from the group consisting of substituted or unsubstituted hydrocarbylene, and substituted or unsubstituted heterohydrocarbylene.

45. The polymer electrolyte of claim 44 wherein R<sup>1</sup> is a solvent moiety derived from tetrahydrofuran, dimethylformamide, dimethylacetamide, N-methylpyrrolidone, dimethylsulfoxide, sulfolane, acetonitrile and propylene carbonate.

46. The polymer electrolyte of claim 44 wherein R<sup>1</sup> is a heterohydrocarbyl solvent moiety having the formula -O(CH<sub>2</sub>CH<sub>2</sub>O)<sub>b</sub>CH<sub>3</sub>, wherein b ranges from about 1 to 6.

47. The polymer electrolyte of claim 40 wherein the polymer further comprises a metal salt.

48. The polymer electrolyte of claim 47 wherein the metal salt is selected from the group consisting of transition metals, alkali metals, alkaline earth metals, or a combination thereof.

49. A covalently cross-linked polymer electrolyte, the polymer electrolyte comprising amine groups in the polymer backbone and labile protons therein.

50. The polymer electrolyte of claim 49 wherein a ratio of protons to nitrogen atoms ranges from about 0.2:1 to about 0.8:1.

51. The polymer electrolyte of claim 49 wherein the polymer is swollen with a protic solvent.

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52. The swollen polymer electrolyte of claim 51 wherein the swelling solvent is selected from the group consisting of water, triflic acid, acetic acid, phosphoric acid, and mixtures thereof.

53. A fuel cell comprising:  
a proton-conducting, covalently cross-linked polymer electrolyte membrane, the polymer comprising amine groups in the polymer backbone and labile protons;

5 an anode in contact with a first side of the membrane; and,  
a cathode in contact with a second side of the membrane, which is opposite said first side.

54. The fuel cell of claim 53 wherein the polymer electrolyte is a linear or branched, substituted or unsubstituted poly(alkylamine).

55. The fuel cell of claim 54 wherein the polymer electrolyte is a substituted or unsubstituted, branched poly(ethylenimine) or poly(propylenimine).

56. The fuel cell of claim 54 wherein the polymer electrolyte is a substituted or unsubstituted, linear poly(ethylenimine) or poly(propylenimine).

57. A battery comprising a negative electrode, a positive electrode, and an ionically conductive polymer electrolyte disposed there between and in contact therewith, wherein said polymer electrolyte is covalently cross-linked and comprises amine groups in the polymer backbone and a metal salt.

58. The battery of claim 57 wherein the polymer electrolyte is a linear or branched, substituted or unsubstituted poly(alkylamine).



59. The battery of claim 58 wherein the polymer electrolyte is a substituted or unsubstituted, branched poly(ethylenimine) or poly(propylenimine).

60. The battery of claim 58 wherein the polymer electrolyte is a substituted or unsubstituted, linear poly(ethylenimine) or poly(propylenimine).

61. A gradient battery comprising a continuous, covalently cross-linked poly(amine) film, the polymer film comprising metal ions, a negative electrode region, a positive electrode region, and an electrolyte region dispose there between which, during charge or discharge of the battery,  
5 enable the passage of metal ions or protons from one electrode to the other.

62. The battery of claim 61 wherein the polymer electrolyte is a linear or branched, substituted or unsubstituted poly(alkylamine).

63. The battery of claim 62 wherein the polymer electrolyte is a substituted or unsubstituted, branched poly(ethylenimine) or poly(propylenimine).

64. The battery of claim 62 wherein the polymer electrolyte is a substituted or unsubstituted, linear poly(ethylenimine) or poly(propylenimine).

65. The battery of claim 62 wherein the electrolyte is a covalently cross-linked polymer single ion electrolyte, the polymer electrolyte comprising amine groups in the polymer backbone and an ion pair, one member of the pair being covalently attached to the polymer backbone and the other being  
5 capable of diffusing through the polymer electrolyte upon the application of an electric field.

5 66. A covalently cross-linked polymer single ion electrolyte, the polymer electrolyte comprising amine groups in the polymer backbone and an ion pair, one member of the pair being covalently attached to the polymer backbone and the other being capable of diffusing through the polymer electrolyte upon the application of an electric field.

• 67. The single ion electrolyte of claim 66 wherein the polymer is a linear or branched, substituted or unsubstituted poly(alkylamine).

68. The single ion electrolyte of claim 67 wherein the polymer is a substituted or unsubstituted, branched poly(ethylenimine) or poly(propylenimine).

69. The single ion electrolyte of claim 67 wherein the polymer electrolyte is a substituted or unsubstituted, linear poly(ethylenimine) or poly(propylenimine).

70. An electrolytic cell comprising:

an anode,

a cathode, and

5 a covalently cross-linked polymer single ion electrolyte, the polymer electrolyte comprising amine groups in the polymer backbone and an ion pair, one member of the pair being covalently attached to the polymer backbone and the other being capable of diffusing through the polymer electrolyte upon the application of an electric field.

71. The electrolytic cell of claim 70 wherein the polymer is a linear or branched, substituted or unsubstituted poly(alkylamine).

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72. The electrolytic cell of claim 71 wherein the polymer is a substituted or unsubstituted, branched poly(ethylenimine) or poly(propylenimine).

73. The electrolytic cell of claim 71 wherein the polymer electrolyte is a substituted or unsubstituted, linear poly(ethylenimine) or poly(propylenimine).

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